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STUDY OF THE $Sm^{149}(n,\alpha)Nd^{146}$ REACTION WITH THERMAL NEUTRONS

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ABSTRACT

The cross section for the $Sm^{149}(n,\alpha)Nd^{146}$ reaction at thermal neutron energies was measured by observing the alpha-particle spectrum of natural Sm and isotopically enriched Sm^{149} targets in the thermal column of a reactor. Alpha-particle groups were observed at 8.72 and 9.12 Mev corresponding to transitions to the first excited state and ground state of Nd^{146}. The 2200-m/sec cross sections (σ_o) for the two groups are 121±15 mb and 22±10 mb respectively. The 4- resonance at 0.0967 ev accounts for most of the cross section to the first excited state, but cannot contribute to the population of the ground state. It is postulated that the population of the ground state arises from a contribution of a bound 3- state in Sm^{150}. Calculated values for the n,α cross section are compared with the experimental results.

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I. Introduction

Although the n,α reaction at thermal neutron energies is energetically

possible for several nuclides throughout the periodic table, the probability

for alpha-particle emission from the capturing state of the compound nucleus

is usually very small. Only for those reactions in which the energy available

for α-particle emission is comparable with the Coulomb-barrier height can this

mode of decay compete favorably with γ-ray emission. The thermal neutron n,α

cross sections which have been reported are for nuclides with $Z \leqslant 30$.[1]

In the region of the rare earth elements, there are a few stable nuclides

for which the thermal neutron n,α reaction is exothermic by approximately

9 Mev. This is still substantially below the Coulomb barrier. One of these

nuclides, Sm^{149}, has a large thermal neutron capture cross section (σ_o = 39,900

barns),[2] so that the possibility of observing the rare alpha-decay mode from

the capturing state is considerably enhanced. The purpose of the present

investigation was to search for this alpha-decay branch by studying the α-

particle spectrum of a Sm target in a thermal neutron flux. In Sm^{149}, the

α-particle spectrum can give information on the spin and parity of the capturing

state as well as the Q value of the reaction.

II. Experimental Details

A. Targets

Targets consisted of natural Sm and samples isotopically enriched in Sm^{149}. The isotopic analysis of the enriched Sm^{149} sample is given in Table I. The targets (approx 100 $\mu g/cm^2$) were deposited homogeneously on a 5-mm-thick Al backing. They were prepared by spreading out an alcohol solution of $Sm(NO_3)_3$ on the Al foil over an area of 25 cm^2 and allowing it to dry under a heat lamp. The foil was then heated to 400° C to drive off the alcohol and convert the nitrate to an adhering deposit of the oxynitrate. Targets 4 cm^2 in area were cut from the homogeneously deposited portion of the sample. Mixed Sm-Li targets, which were used for cross-section measurements, were prepared by using an alcohol solution of $Sm(NO_3)_3$ and Li_2O and depositing in the same manner. These mixed targets were cut from different parts of the deposit in order to determine whether the Sm/Li ratio was constant over the entire deposit. The experimental results did not indicate any fluctuation of the Sm/Li ratio for these targets.

B. Target Assembly and Irradiation Conditions

The targets were mounted on a circular lucite plate which fitted on the front end of the target assembly. The plate was connected to a drive shaft which extended outside the thermal column of the reactor when the assembly was in the irradiating position. The drive shaft was manually driven to position different targets in front of the α-particle detector while the reactor was in operation. Measurements were made in air with a distance of 3 mm between the target and detector.

The target assembly was irradiated in the thermal column of the Puerto Rico Nuclear Center Research Reactor at a position separated from the lead face of the core by 1.6 meters of graphite. The temperature of the graphite

moderator was 32° C.[3] The Cd ratio for a "1/v" detector at this position was not measured, but should be much greater than 1000 on the basis of measurements at an equivalent position with the Livermore Pool Type Reactor (LPTR).[4]

Alpha-particle spectra were obtained with a gold-surface-barrier detector connected to standard electronics and a 512-channel pulse-height analyzer. The detector is made from n-type 1800-ohm-cm silicon and operated at a reverse bias of 30 v. The active surface area is 0.8 cm^2. Alpha-particle energy calibration was obtained by using a Th^{228} source. The α-particle energies used were Th^{228}, 5.40 Mev; Ra^{224}, 5.68 Mev; Em^{220}, 6.28 Mev; Po^{216}, 6.77 Mev; Po^{212}, 8.78 Mev.[5]

C. Background Measurements

Several experiments were performed in order to determine the contribution of the non-Sm part of the targets to the background in the region of interest (6 to 12 Mev). Alpha-particle spectra were obtained with lucite, Li_2O, and Al targets in the thermal neutron flux. No contribution to the background above 6 Mev could be observed for any of these materials. The background counting rate above 6 Mev was approximately 0.1 count/min. Alpha-particle measurements of the natural Sm and enriched Sm^{149} targets at zero flux failed to show any contribution to the background from U or Th contamination of the samples.

D. Cross-Section Measurements

By use of the mixed Sm-Li targets, the counting rate of the α particles from the $Sm^{149}(n,\alpha)$ reaction was compared with the counting rate of tritons from the $Li^6(n,\alpha)T^3$ reaction. This procedure made it possible to obtain the

$Sm^{149}(n,\alpha)$ cross section without evaluating the neutron flux and counting geometry. The intensity of the 2.74-Mev T^3 peak was used in the analysis because the resolution of the peak was less affected by sample thickness than was that for the 2.05-Mev α-particle group. The energy of the latter group was also appreciably attenuated by the 3 mm of air between the target and detector.

The counting rate of the two n,α reactions was obtained from the particle spectra by integrating the counting rate over the peaks. In both cases the integration was terminated at an energy at which the counting rate was less than 5% of the peak height. A background subtraction was made by using the background observed for a lucite target at the same flux level. The cross section was obtained from the expression

$$\hat{\sigma}_{n,\alpha}(Sm^{149}) = \frac{C_{Sm}}{C_{Li}} \times \frac{\sigma_{o_{n,\alpha}}(Li^6) \times i_6 \, f}{i_{149}} \tag{1}$$

where

$\hat{\sigma}_{n,\alpha}(Sm^{149})$ = the effective thermal neutron n,α cross section for Sm^{149} in a Maxwellian neutron flux (t $\approx$ 305°K),

$\sigma_{o_{n,\alpha}}(Li^6)$ = the 2200-m/sec n,α cross section for Li^6 = 945 barns,[1]

C_{Sm} = net counting rate from the $Sm^{149}(n,\alpha)Nd^{146}$ reaction,

C_{Li} = net counting rate from the $Li^6(n,\alpha)T^3$ reaction,

f = atom ratio of natural Li to Sm,

i_{149} = isotopic abundance of Sm^{149},

i_6 = isotopic abundance of Li^6 = 0.0742.[6]

Because of a resonance at thermal energies, neutron absorption in this region does not follow the "1/v" law for Sm^{149}. The effective absorption cross section for a Maxwellian distribution of neutron energies, when divided by a factor g, yields the cross section for 2200-m/sec neutrons (σ_o).[7,8] This factor, which is dependent upon neutron temperature, represents the departure of the absorption cross section from 1/v dependence. It has been calculated by Pattenden over a range of neutron temperatures for Sm^{149}.[2] The neutron temperature for this experiment was taken as equal to the temperature of the thermal column, $305°K$. At this temperature, g is 1.74. The variation of g with neutron temperature is not large enough to introduce a significant error if $305°K$ does not correspond to the actual neutron temperature. The contribution of epithermal neutrons to the cross section is expected to be negligible because of the high Cd ratio at the irradiating position.

III. Results

Preliminary measurements were made with thick Sm targets at fluxes up to 10^{10} $n/cm^2/sec$ in order to determine whether any high-energy α particles could be detected. Counting rates on the order of hundreds of counts per min were observed at energies greater than 6 Mev, indicating that the n,α cross section could be easily measured. Subsequent experiments were performed with much thinner samples (100 $\mu g/cm^2$) and at a lower neutron flux (approx 3×10^8 $n/cm^2/sec$) in order to obtain good α-particle resolution.

The neutron-induced particle spectra of natural Sm and of enriched Sm^{149} targets both show groups at 8.72 and 9.12 Mev (Fig. 1). The intensity of the higher-energy group is approximately one-tenth the intensity of the lower-energy group. An upper limit of one-tenth the intensity of the 8.72-Mev group was established for the existence of lower-energy groups.
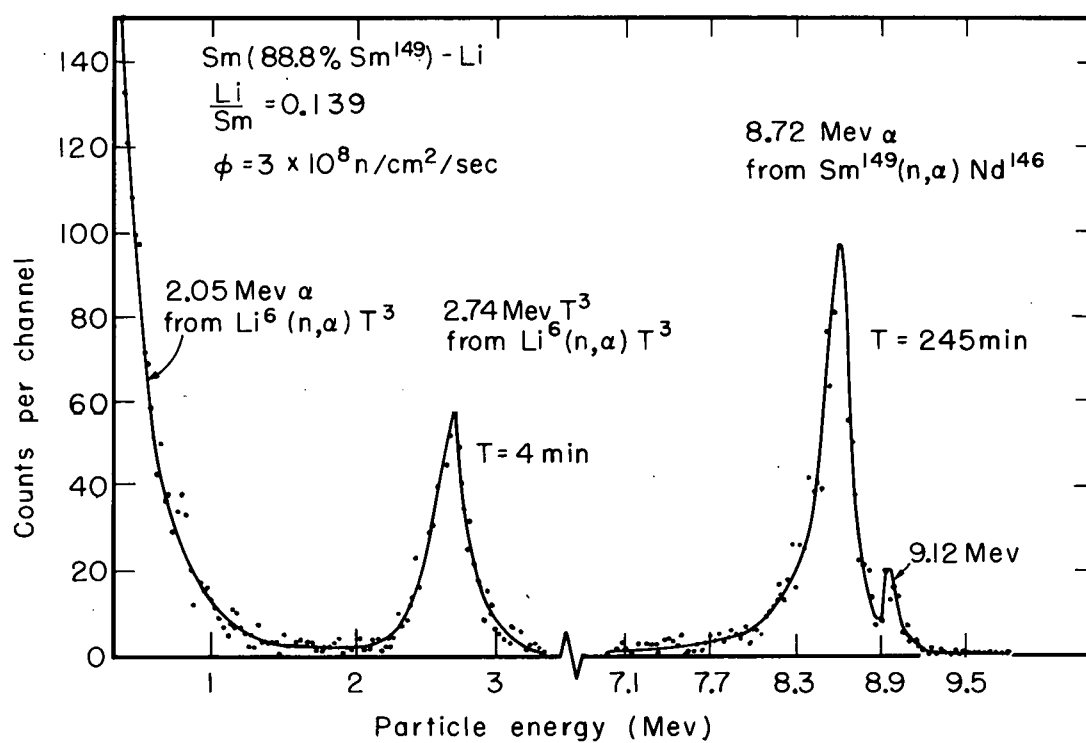
Fig. 1. Particle spectra for a mixed Sm-Li target in a
thermal neutron flux.

To determine the identity of the particles, a thin Al foil of known thickness was placed between the detector and target. The observed degradation of the energy of the 8.72-Mev group corresponded to that expected for α particles of that energy.

The cross section results are given in Table II.

IV. DISCUSSION

A. Energetics and Decay Scheme

The Q value for the $Sm^{149}(n,\alpha)Nd^{146}$ reaction calculated from atomic mass differences is 9.47 ± 0.15 Mev.[9,10] This agrees well with the Q measured for the higher-energy alpha group (9.38 ± 0.04 Mev). The Q value for the more prominent group is 8.96 ± 0.04 Mev, which is consistent with that expected for a transition to the first excited state of Nd^{146}, a 2+ state lying 455 kev above the ground state.[11] This agreement with the Q values from atomic mass data suggests that most of the alpha decay takes place from the capturing state of Sm^{150} and not from intermediate states in Sm^{150} populated by γ-ray emission from the capturing state. A proposed decay scheme is given in Fig. 2.

Neutron capture for Sm^{149} in the region of thermal energies is dominated by a resonance at 0.0967 ev.[12] The spin of the capturing state is 4, the result of an I+1/2 addition[13,14,15] of an s-wave neutron to the nuclear spin of Sm^{149} (I = 7/2).[16] The parity of the ground state of Sm^{149} has not been experimentally established. However, shell-model theory favors an odd parity, with the unpaired neutron occupying an $f_{7/2}$ level.[17] The parity of the capturing state would also be expected to have an odd parity for s-wave neutron capture. Alpha-particle emission from this state can take place with $\ell = 3$ and $\ell = 5$ waves to the first excited state of Nd^{146}, but the ground-state transition is forbidden by parity conservation. This state is presumably responsible for the
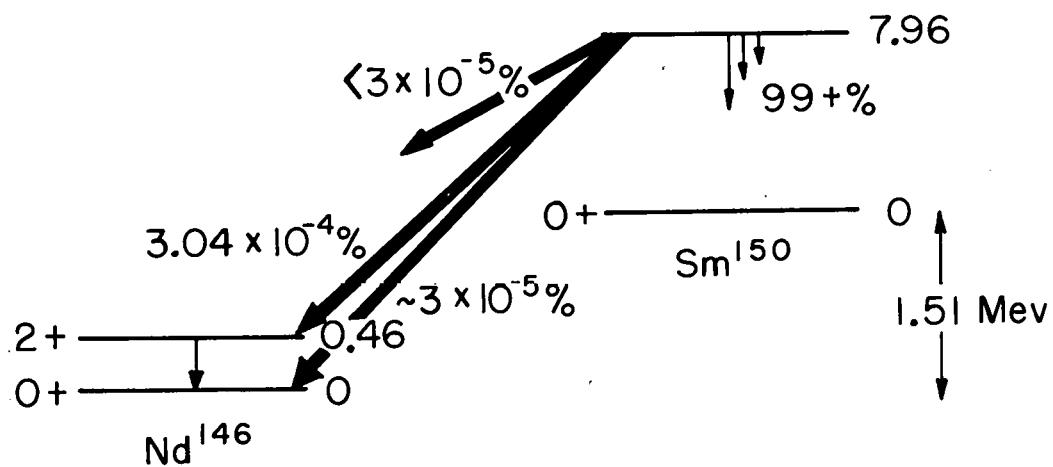
Fig. 2. Proposed partial decay scheme of the s-wave
         neutron capturing state of Sm^{150}.

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major part of the n,α cross section to the first excited state of Nd^{146}.

However, no transitions to the ground state should have been observed if the

0.0967-ev resonance were the only contributing state. This suggests a contri-

bution from an I-1/2 or 3- state which can alpha decay to the Nd^{146} ground

state by an ℓ = 3 wave. A second resonance occurs at 0.87 ev, but it is also

formed by an I + 1/2 addition of the neutron.[15] The 1/v component of higher-

energy resonances in the thermal region is too small to account for the observed

n,α cross section. A possible source of a contributing 3- state may be found

in the bound levels of Sm^{150} lying just below the neutron separation energy.

Pattenden, in fact, observed that there appeared to be an additional 1/v

component of a magnitude equal to $760/\sqrt{E}$ ev barns in the cross-section data

for Sm^{149} at neutron energies below the 0.0967-ev resonance.[2] If this were

due to the contribution of a bound 3- level, it would provide an explanation

for the appearance of the ground-state alpha decay in the $Sm^{149}(n,\alpha)Nd^{146}$

reaction.

In the calculation of the n,α cross section discussed below, the

assumption is made that a bound 3- state contributes to the thermal neutron

cross section.

B. Comparison of the Experimental Cross Section with Calculated Values

Calculations of the n,α cross section for a large number of nuclides

have been made by Griffioen and Rasmussen.[18] The essential features of their

analysis are retained in the calculation presented here. The calculation of

the cross section for the n,α reaction to the 2+ state of Nd^{146} requires a

knowledge of $\Gamma_\alpha^I(J,\ell)$ for the capturing states, where I is the spin of the

final state, J is the spin of the initial capturing state, and ℓ is the

angular momentum carried off by the α particle. The contributing alpha widths

for each of the states are given in the expression

$$\Gamma_\alpha^{2+}(\text{total}) = \Gamma_\alpha^{2+}(4,3) + \Gamma_\alpha^{2+}(4,5) + \Gamma_\alpha^{2+}(3,1) + \Gamma_\alpha^{2+}(3,3) + \Gamma_\alpha^{2+}(3,5),$$

(2)

$$\Gamma_\alpha^{0+}(\text{total}) = \alpha^{0+}(3,3).$$

(3)

The alpha width for a given value of ℓ was obtained from[19]

$$\Gamma_\alpha^{I}(J,\ell) = \frac{D}{2\pi} T_\ell,$$

(4)

where D is the average level spacing for states near the capturing states of the same spin and parity, and T_ℓ is the transmission probability for an ℓ-wave α particle.

Transmission probabilities for Q_α = 8.96 and 9.38 Mev were calculated for the different ℓ waves by using a computer program written by Rasmussen,[20] which calculates T_ℓ for a nuclear potential including the real part of the optical-model potential.[21] From these values of T_ℓ, $\Gamma_\alpha^{I}(J,\ell)$ was calculated from Eq. (4) using a value of D = 6.6 ev,[22] which was obtained from an analysis of the neutron excitation-function data for Sm^{149}. The results are given in Table III.

The n,α cross section for ℓ-wave alpha emission from the state of spin J to the state of spin I was obtained from the n,γ cross section by using the expression

$$\hat{\sigma}_{n,\alpha}^{I}(J,\ell) = \frac{\Gamma_\alpha^{I}(J,\ell)}{\Gamma_\gamma^{I}(\text{total})} \times \hat{\sigma}_{n,\gamma}^{I}(\text{total}),$$

and

$$\hat{\sigma}_{n,\alpha}^{I}(\text{total}) = \frac{\Gamma_\alpha^{I}(\text{total})}{\Gamma_\gamma^{I}(\text{total})} \times \hat{\sigma}_{n,\gamma}^{I}(\text{total}),$$

(6)

which is valid if the n,γ cross section and the capture cross section are essentially the same and if the total γ-ray width is much larger than the width for any other mode of decay. The contribution of the proposed 3- bound state to the n,γ cross section was treated in the following manner. The 2200-m/sec cross section, σ_0, was multiplied by g (1.74) to obtain the effective cross section, $\hat{\sigma}_{n,\gamma}^{3+4}(\text{total})$, for a Maxwellian distribution of neutrons of temperature $305^\circ K$. A contribution of 4800 barns $\hat{\sigma}_{n,\gamma}^{3}(\text{total})$ from the 3- state was subtracted from the total on the assumption that the n,γ cross section is given by $760/\sqrt{E}$ ev barns.[2] It was assumed that $\Gamma_\gamma^3(\text{total})$ for the 3- state is the same as for the 4- resonance at 0.0967 ev. [The three lowest-energy resonances of Sm^{149} all have approximately the same $\Gamma_\gamma(\text{total})$[1]]. A value $\Gamma_\gamma^4(\text{total}) = 0.058$ ev was used.[2]

Calculated values of $\hat{\sigma}_{n,\alpha}^{I}(J,\ell)$ are given in Table IV together with $\hat{\sigma}_{n,\alpha}^{I}(\text{total})$. The total calculated effective cross section for the n,α reaction to the 2+ state $[\hat{\sigma}_{n,\alpha}^{2}(\text{total})]$ is 132 mb, while the experimental value is 211 ± 27 mb. The calculated cross section for n,α to the ground state is 31 mb, compared to the experimental value of 22 ± 10 mb. Since the calculation is expected to provide at best only an order-of-magnitude value for the n,α cross section, the agreement is surprisingly good.

One of the interesting features of the n,α reaction with thermal neutrons is the use of an experimentally determined value for D in the calculation of the alpha width. This is not possible for the ground-state alpha emitters where the idea of an average level spacing for states of the same spin and parity is somewhat ambiguous. The results obtained for the $Sm^{149}(n,\alpha)$ cross section give some experimental support to the relation between D and the period of motion of the nucleus at high excitation energy.[19]

The thermal neutron n,α cross section may provide a means of studying properties of excited states in the rare earth region in much the same manner as with ground-state alpha emitters in the heavy elements. Population of excited states at energies exceeding 1 Mev should be possible with the n,α reaction in the rare earth region because T_ℓ varies less rapidly for large E_α. As an example, the $Sm^{149}(n,\alpha)$ cross section to the 1.21-Mev level in Nd^{146} is expected to be approximately 5% the cross section to the first excited state.

Table I. Isotopic analysis of enriched Sm^{149} sample.[a]

Mass	Isotopic Abundance (%)
147	1.0 ± .05
148	4.0 ± .1
149	88.8 ± .1
150	3.5 ± .1
152	1.9 ± .05
154	0.9 ± .05

[a] Obtained from Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Table II. Cross section results

Sample	Q_α (Mev)	$\hat{\delta}_{n,\alpha}$(Sm^{149}) (mb)	$\sigma^o_{n,\alpha}$(Sm^{149}) (mb)
Natural Sm 88.8% Sm^{149}	9.38 ± 0.04	15 ± 10	15 ± 10
88.8% Sm^{149}	9.38 ± 0.04	22 ± 10	22 ± 10
Best value		22 ± 10	22 ± 10
Natural Sm 88.8% Sm^{149}	8.96 ± 0.04	235 ± 54	135 ± 31
Sample No. 1	8.96 ± 0.04	202 ± 18	116 ± 10
Sample No. 2	8.96 ± 0.04	221 ± 20	127 ± 11
Average (best value)		211 ± 27	121 ± 15

Table III. Alpha-particle transmission probabilities and alpha-decay widths for the $Sm^{149}(n,\alpha)Nd^{146}$ reaction.

Q_α (Mev)	ℓ	T_ℓ	Γ_α
8.96	1	1.89×10^{-7}	1.99×10^{-7}
8.96	3	7.33×10^{-8}	7.70×10^{-8}
8.96	5	1.36×10^{-8}	1.43×10^{-8}
9.38	3	3.48×10^{-7}	3.66×10^{-7}

Table IV. Calculated n,α cross sections

Transition	Alpha-particle ℓ wave	$\hat{\sigma}_{n,\gamma}(Sm^{149})$ (barns)	$\Gamma_\alpha/\Gamma_\gamma$	$\hat{\sigma}_{n,\alpha}(Sm^{149})$ (mb)
$4- \rightarrow 2+$	3	64,700	1.33×10^{-6}	86.0
	5	64,700	2.47×10^{-7}	16.0
$3- \rightarrow 2+$	1	4,800	3.43×10^{-6}	16.4
	3	4,800	1.33×10^{-6}	6.4
	5	4,800	2.47×10^{-7}	1.2
Total to 2+				126.0
			Experimental	211 ± 27
$3- \rightarrow 0+$	3	4,800	6.31×10^{-6}	30.3
			Experimental	22 ± 10

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